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Atmospheric Chemistry and Physics Discussions

Interactive comment on "MAX-DOAS measurements in southern China: 1. automated aerosol profile retrieval using oxygen dimers absorptions" *by* X. Li et al.

X. Li et al.

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Reply to both referees

We thank both referees for their comments on our manuscript. Both referees suggested to remove the two parameter fit and to change the number of free parameters of the A5 fit from 4 to 3. These changes required a number of recalculations, updates in the table, new figures, and major modifications in the text. We also included more information about measurement errors and we calculated a humidity corrected aerosol scattering time series. We found some shortcomings in the calculations of the errors

and the aerosol layer height. These were corrected in the revised version. The number of figures was reduced from 9 to 7 as suggested by the referees' reports.

Reply to Referee #1

We thank referee #1 for the positive and critical review of our manuscript. In the following we will answer his/her questions step-by-step and explain how we changed the revised version of the manuscript.

Comment: It is currently difficult to understand how the measurement errors are considered in the retrieval. I recommend that the authors expand this topic to make it easier for the reader to follow their approach. For example, the manuscript does not present the typical statistical and systematic error of the O4 DSCD. The authors should also explain the weighing in equation 9 in more detail, i.e. where do the weights come from, etc. In addition, Figure 3, 8, and 9 need error bars.

***Answer**: The error of the O4 DSCD is composed by two parts, statistical error and systematic error. The statistical error represents the precision of the measurement and is derived from the DOAS fit, i.e. the random error of the linear least square fit. The systematic error describes the accuracy of the measurement and is mainly determined by the accuracy of the reference absorption cross sections which we consider 10%, 3%, 4%, 8%, and 5% for O4, NO2, O3, BrO, and HCHO, respectively. Since the systematic error of the cross section is the same for all data point, it is not considered in the fitting procedure. Adding error bars to the Figure 3 will overwrite the major content of the figures. However, we plotted error bars in Figures 4, 5, and 6, as suggested by the referee. The weights are deduced from measurement error (precision) of the O4 DSCD. In addition we changed the text in order to address this point.*

Comment: What is the reason for including the results of case A4 in the manuscript? It seems to me that using an approach that is based on an incorrect description of the

atmosphere, i.e. omitting a variable boundary layer height, is destined to fail. A least squares fit based on a physically incorrect model will yield incorrect results, even if they seem statistically sound. The manuscript would not suffer from omitting case A4 completely.
***Response**: As suggested by both referees we removed the two parameter fit and changed the number of free parameters of the A5 fit from 4 to 3. The text, tables, and figures were changed accordingly.*

Comment: The drying of the air analyzed by the nephelometer could cause a bias in the comparison of the in-situ vs. the MAX-DOAS extinction coefficients. This bias needs to be discussed in more detail. Showing the relative humidities during the experiment and presenting a back-of-the-envelope calculation of the change of the aerosol size and extinction coefficient during the drying process would allow an assessment of this bias and improve the comparison between nephelometer and MAX-DOAS data.
***Response**: We introduced a new, humidity corrected set of the nephelometer data. The correction procedure is described in Eq. 7 and is based on parameters published by Liu et al. (2008). The corrected data are based on humidity recorded on the BG site. The corrected data are included in Figure 5 and are used for the correlations in Figures 6A and 6B. The text and figure captions are changed accordingly.*

Comment: Figure 8 shows considerable variations of the boundary layer height in the afternoon, i.e. the retrieved BLH drops suddenly at 15:30 and at 17:30. I am not aware of a mechanism that could lead to a reduction of BLH on these short timescales. Are these results statistically significant or is there another cause for such a rapid change in BLH? Averaging the data over 9 days seems to smooth the BLH variation, but this approach seems somewhat arbitrary. It is unclear if the averaging just reduces statistical uncertainties or if there is a problem with the measurement/retrieval process for individual data points. These issues need to be addressed to give the reader confidence that the methods described in the manuscript are sound.
***Response**: Thanks for the comment which put our attention on Figure 8 (which data is*

now included in Figure 5) which had an error being corrected in the updated version. In the correct data in Figure 5, does not show sudden drops at 15:30 and 17:30 on 2006-7-24. However, some some sudden variations pesist (e.g. at 15:30 on 2006-7-21) and reflect either a possible change in the air mass or the limitation of the method. We discuss that in the revised version. Figure 7a (in the revised version, was Figure 9) now show the average diurnal profile of the aerosol layer height together with the individual data to demonstrate the variability of the retrieved parameter. At this point we think that a error bar based on the standard deviation could be misleading. The same kind of analysis is provided vor E_0.

Reply to Referee #2

We thank referee #2 for his/her detailed and critical review of our manuscript. In the following we will answer his/her questions step-by-step and explain how we changed the revised version of the manuscript.

Comment: The paper by Li et al. reports on observations of aerosol parameters using ground based MAX-DOAS measurements in the polluted environment of Pearl River Delta region 50 km north of Guangzhou. The study focuses on two major topics: 1. The concept for the retrieval of the aerosol extinction and the height of the boundary layer using O4 absorption measurements in the UV is described. 2. For a limited data set (nine days) the results of the aerosol retrieval are compared to ground-based nephelometer measurements of aerosol properties. The usage of MAX-DOAS measurements to derive aerosol properties has already been shown in previous studies (e.g. Heckel et al., 2005, Wittrock et al., 2004, and in more detail in Irie et al., 2008, new paper in ACPD). But the technique is in general novel and validated data sets in particular for regions with high aerosol load are scarce. For that reason the study is clearly suitable for publication when the authors address for major revisions/corrections

as detailed below. Since the paper is more of technical nature and the scientific outcome limited it might be worthwhile to publish the paper in the new journal "Atmospheric Measurement Techniques (AMT)".
***Response**: This paper is part of a special section on PRD campaign 2006 (ACPD: http://www.atmos-chem-phys-discuss.net/special_issue101.html; ACP: http://www.atmos-chem-phys.net/special_issue164.html) and fits in to the frame work of the papers submitted and being prepared for this section. In addition it also provides input parameters, i.e. BLH for the interpretation of other ground based measurements at the site where the MAX-DOAS was located. In the revised version we put more emphasis on the papers which were mentioned by the referee.*

Comment: I agree with referee 1, that more information on the uncertainties of data presented here is needed, in particular, how the O4 error bars are considered in the retrieval. What's also missing is a more detailed statement on the errors of the nephelometer measurements.
***Response**: Part of the answer was given to referee #1. The error of the precision of the DSCDs was used as weight in Eq. 6. The nephelometer data are described in detail in the paper by Garland et al. (2008). We added a statement about the nephelometer error (10 %) in the text.*

Comment: I am quite critical about the argument on the maximum number of retrieval parameters (page 17669, last paragraph) for several reasons: On one hand measurements for different elevation angles are not independent from each other (depending on actual meteorological conditions), on the other hand each retrieval step comprises more than one scan and hence different azimuth and solar zenith angles (which increases the degrees of freedom). In principle the authors are right, that the information content from this type of measurements is limited. But the real number of possible parameters (degrees of freedom) to be retrieved changes dramatically with actual conditions. Therefore more advanced retrieval methods like optimal estimation are able to characterize the degrees of freedom for each data point. Friess et al. have made an

excellent study on theoretical aspects of the aerosol retrieval from O4 measurements and came out with a number of less than 2 (degrees of freedom) for a retrieval taking into account only the O4 absorption in the UV. The authors should change their argumentation by addressing these issues.
***Response**: As described by Friess et al. (2006), only 2 pieces of information (degree of freedom) on the aerosol extinction below 500 m can be derived when using merely the O4 absorption at a single wavelength. Different from Friess et al. (2006), we have 6 elements (i.e. elevation angles) instead of 4 in the measurement vector, which will certainly increase the degree of freedom of the retrieval. Moreover, we analyzed the averaging kernels for the parameters we defined in order to further check the degree of freedom of the retrieval. We found the peak value of nearly unity of τ, F and H are located at the corresponding variable. This suggests that the information of TAU, F and H can be derived from the MAXDOAS measurement. But the averaging kernel of the scaling height ξ is near zero indicating the small sensitivity of MAXDOAS measurements to H0. This clearly justifies the the use of a 3 parameter fit.*

Comment: Description of the extinction profile (Section 3): To my knowledge the extinction profile in polluted areas is highly variable with altitude and a well-mixed boundary layer unlikely (see e.g. Ansmann et al., 2005, High aerosol load over the Pearl River Delta, China, observed with Raman lidar and Sun photometer, GRL). The authors should consider this in their error discussion.
***Response**: We are not in agreement with the referee's comments here. As Figure 4 in the paper of Ansmann et al. (2005) shows, the aerosol extinctions in each layer below 1 km are not statistically different from each other. The assumption of a well-mixed aerosol layer near ground should be valid. However, to be more straightforward, it is better to change the term boundary layer to aerosol layer.*

Comment: Why the authors introduce the scaling height for the aerosol in the free troposphere as an additional retrieval parameter? Several studies before (e.g. Friess et al., Wittrock et al., 2004, and much more) have shown, that the sensitivity of MAX-

DOAS observations to higher altitudes is very small. This is in particular the case for a high aerosol load and in the UV spectral range. If not applying a full profile retrieval like optimal estimation I would expect the best results by using a three-parameter model (A5 without scaling height in this study).
***Response**: We agree with the referee that the small sensitivity of MAXDOAS observations on the aerosol at higher altitudes and thus H0 (as discussed above). For the revised version we recalculated the entire data set with a scaling height of $\xi = 5\,km$*

Comment: For similar reasons as referee 1 I am sceptical about the benefit of the two-parameter model A4. As also pointed out by the authors this model gives reasonable results for high aerosol load only.
***Response**: As suggested by both referees we removed the scenario A4 from the manuscript. The text, tables, and figures were changed accordingly.*

Comment: Title: Since the information output of the presented data set is quite limited, I suggest to change the title to ... aerosol retrieval using ... . The word "profile" implies much more than a few values for the aerosol properties and only one of them being validated.
***Response**: We changed the title to "MAX-DOAS measurements in southern China: Automated aerosol retrieval and validation using ground-based in-situ data" .*

Comment: Abstract: The second sentence is misleading. The sampling for different elevation angles is done sequentially not simultaneously. Furthermore the O4 absorption is analysed between 352 and 390 nm which comprises two absorption bands at 360 and 380 nm, respectively.
***Response**: We reworded the abstract based on the above comments: "Here we show that the O_4 (O_2 dimer) absorptions at 360 nm and 380 nm can be used to retrieve the aerosol extinction and the height of the boundary layer.*

Comment: Introduction: I recommend to redraft the whole section. In general, I would expect a more detailed discussion on other papers showing the practical possibilities

to retrieve aerosol properties from MAX-DOAS O4 measurements. There is no need to spend time and space to explain the basic concept behind DOAS or MAX-DOAS since this has been done several times in other publications (in a more precise way, see below). It is also not clear to me, why the author describe in detail properties of the used radiative transfer model. A lot of the information given here is needless at this point and should be shifted to section 3.
***Response**: We removed several paragraph from the introduction. The description of the RTM is now in Section 3, as suggested. The RTM was described in more detail since the thesis of Deutschmann was missing at the time of the submission. We now included a link to the thesis and the program.*

Comment: - line 11: (Platt and Stutz, 2008 and references therein), - line 14: remove "profiles", - line 16 and below: add for NO2 (Wittrock et al., 2004, Brinksma et al., 2008, Irie et al., 2008), for HCHO (Heckel et al., 2005), for CHOCHO (instead of glyoxal) (Wittrocket al., 2006), - line 18: change "developed" to "described"
***Response**: all changed in the revised version.*

Comment: page 17663: The "idea" of DSCDs is not only introduced for the description of measurements focusing on the troposphere. The DSCD is the standard outcome for DOAS observations from the ground, since there is no possibility to retrieve the SCD without any further assumptions. Please change all equations accordingly.
***Response**: In the text we state that DSCDs are used for MAX-DOAS of tropospheric species. We do not claim that this is used exclusively. However, the most of the introduction has been changed to meet the suggestions of the reviewer. We hope, that the current, shortened version is much clearer for the reader.*

Comment: page 17664: "Deutschmann": The authors should refer only to publications or thesis which are available to all potential readers (e.g. online with a fixed link).
***Response**: The thesis of Deutschmann can now be downloaded from the web site of McArtim. A link is provided in the revised version.*

Comment: page 17666, line 6: at azimuth. ? word missing? In addition, the measured O4 towards the sun is not always lower than for the other directions. For higher solar zenith angles this is the other way round.
***Response**: It should be ... at azimuth back to the sun. Text was changed accordingly.*

Comment:Experimental: What is the field of view of the instrumental setup? What's the impact on the results?
***Response**: The field of view of the telescope is 0.3°. The field of view of the telescope is determined by the size of the fiber and the lens and it was not changed during the period. Within the error of the field of view there is only a marginal influence on the results as we calculated from sensitivity runs of the RTM.*

Comment:- page 17666, line 2: "contains" instead of "containes", - page 17668, line 1: "with adjustments"
***Response**: changed in the revised manuscript.*

Comment: -I0-effect: Is it really needed to take into account this effect? What's the impact on the results?
***Response**: I0-effect was considered along the discussion Platt et al.(1997). The effect on the DSCDs is less than 2 % for the data presented here. A statement was made in the text. At this point we think the use the I0-corrected cross sections is justified.*

Comment: section 2.3: please redraft to avoid too often "our"; Radiative transfer modelling: Not only RTM is described here but the whole retrieval method. Please change the title.
***Response**: We changed the title of the section and rewrote the text accordingly.*

Comment: Page 17670, line 17: "extinction at ground level" ?, that's the same for the whole boundary layer, or not?
***Response**: Yes, it is the same. Clarified in revised version.*

Comment: Page 17672, line 25: really right errors? 1.7 +/- 4.0?
***Response**: During the recalculation (with 3 parameters) we found also a mistake in the error calculation and handling which was removed. The new, smaller error bars are stated in the revised text.*

Comment:Page 17674: Here the authors discuss several reasons on the large deviation between nephelometer and MAX-DOAS in the morning hours. One possible argument is missing: the telescope is pointed towards the east which means towards the sun in the morning. As shown in several studies before the radiative transfer in this case is quite difficult to model due to the forward peak of the aerosol scattering (e.g. Hendrick et al.), consequently the possible error in the modelled O4 column very high.
***Response**: We added a statement about the sensitivity in the discussion.*

Comment: Page 17675: please change "aerosol profiles" to "aerosol properties", see arguments above.
***Response**: was changed in the text.*

Comment: Figure 1: not sure that this figure is needed.
***Response**: We still think this figure helps to provide insight to the data retrieval.*

Comment: Fig 7: definitely useless, all necessary information is given in Fig 8.
***Response**: We agree with referee and removed this figure.*

Comment: Add error bars in particular to figures 8 and 9.
***Response**: We agree with referee and added error bars to Figures 5 and 6. In addition we show the scatter of the data going into the average diurnal profiles (Figure 7).*

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 17661, 2008.